

Ferromagnetic order in $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$

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We present the discovery of ferromagnetism in single crystalline $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ below $T_C = 1.30$ K with a spontaneous magnetic moment of $0.1 \mu_B/\text{Yb}$ along the crystallographic c -axis. This is shown by the huge ac-susceptibility peak, the hysteresis in magnetization as well as by the field-dependent susceptibility and specific heat data. This discovery motivates the reinvestigation of the magnetic order in $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ with $x < 0.27$ and in pure YbRh_2Si_2 under small pressures with field along the c -direction to look for low-lying ferromagnetic order.

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The unambiguous observation of antiferromagnetic (AFM) quantum critical points (QCPs) in heavy-fermion systems has led to an increasing number of theoretical and experimental works in order to understand quantum phase transitions (QPTs) as deeply as their classical counterpart [1, 2]. This work is still in progress, since, although in few cases the standard spin-density-wave model seems to work, in others it does not [3, 4]. The so-called local QCP scenario has been proposed to partially understand some fundamental observations, e.g. the ω/T scaling in $\text{CeCu}_{1-x}\text{Au}_x$ [5], but several other features are still unexplained. Nowadays, it is clear that many of these quantum critical systems are governed by not just one single energy scale and the simple Doniach scenario with one single QCP can not be applied [6]. Proposals concerning a global phase diagram which incorporates the effect of pressure, magnetic field and frustration on the magnetic state as well as on the Kondo effect (including Fermi surface reconstruction) have lead to a new paradigm [7–9]: A system can be tuned across critical points of different nature, e.g. with itinerant or local character. In this global phase diagram the magnetic phase is considered to be antiferromagnetic. However, real systems are more complex and often the dynamical susceptibility is governed by both finite wave vector ($\mathbf{q}=\mathbf{Q}$) and $\mathbf{q}=0$ critical fluctuations [10–12]. Theoretical calculations suggest that when a uniform ($\mathbf{q}=0$) clean itinerant ferromagnet is tuned towards its putative ferromagnetic (FM) QCP, it becomes inherently unstable towards a phase transition of first order or modulated/textured ($\mathbf{q} \neq 0$) phases [13, 14]. Several metals have confirmed this prediction [15–18].

In this respect, the tetragonal heavy-fermion compound YbRh_2Si_2 is a prototype system to study, since it displays AFM order below a very low temperature $T_N = 0.07$ K [19] and is very close to a peculiar AFM QCP [20] where FM critical fluctuations have been observed [10, 21]. At 2.2 mK another phase transition has been observed in magnetization measurements, but its origin is still unrevealed [22]. Pressure or Co substitu-

tion on the Rh site, i.e. $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$, increases T_N as well as the strength of the FM fluctuations [23, 24]. In addition, a second magnetic transition emerges at a temperature $T_L < T_N$ and joins T_N at $x = 0.27$ (≈ 4.3 GPa) [24, 25]. Both phases below T_N and T_L were believed to be AFM since the application of a magnetic field along the magnetic easy axis, i.e. perpendicular to the crystallographic c -axis, depresses the transition temperatures to zero. In this study, we show that the AFM ground state of YbRh_2Si_2 is tuned to a FM one in the Co-substituted compound with $x = 0.27$. This discovery advises to reexamine the magnetic phase diagram of $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ with $x < 0.27$ and of YbRh_2Si_2 under small pressures to look for a possible FM ground state below T_L .

YbRh_2Si_2 is strongly anisotropic in terms of the critical magnetic field B_N which suppresses the AFM order at $T = 0$: $B_{N,\perp c} = 0.06$ T and $B_{N,\parallel c} = 0.66$ T for the magnetic field applied perpendicular and parallel to the c -axis, respectively [26]. The ordered magnetic moment within the AFM phase was determined to be about $2 \cdot 10^{-3} \mu_B$ via muon spin rotation measurements [27]. This small value and the very low T_N make it extremely difficult to determine the magnetic ordering wave vector \mathbf{Q} by means of neutron scattering. Therefore, it is helpful to increase T_N and the magnetic moment by applying hydrostatic pressure, as this usually stabilizes magnetism in Yb systems. This has been investigated in Refs. [23, 28–30]. Moreover, the second phase transition at temperature $T_L < T_N$ emerges above 0.1 K with 0.5 GPa. Comparable behavior can be obtained by iso-electronic substitution of Co on the Rh site (see Fig. 1a). Good agreement concerning the transition temperatures up to about 2 GPa, which corresponds to a Co content $x = 0.12$, was found between the pure system under hydrostatic pressure and the Co-doped samples [24, 25, 31]. However, there are some differences: Pressure experiments have showed that the unit cell c/a -ratio is constant up to the highest applied pressures of 21 GPa [29], while in $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ an increase of c/a has been seen

with decreasing unit cell volume [24]. For small concentrations the change of c/a is linear and amounts to 0.3 % for $x = 0.27$. At $x = 0.27$ (marked by the arrow in Fig. 1a), which corresponds to a hydrostatic pressure of about 4.3 GPa, T_N and T_L appear to merge to a single magnetic transition, while in the pressure experiments by Knebel *et al.* two distinct phase transitions were observed up to 7 GPa [23].

In YbRh_2Si_2 both FM and AFM fluctuations were found to exist [32]. Based on field-dependent specific heat and resistivity experiments under pressure, Knebel *et al.* suggested that the AFM state of YbRh_2Si_2 changes into a FM one above about 7 GPa [23]. Also in $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ the increase of chemical pressure increases the strength of the FM fluctuations. This has been concluded from Curie-Weiss fits performed within a fixed temperature range above T_N , where the Weiss temperature Θ_W increases with increasing x and even switches from negative to positive values at $x = 0.27$ [24]. However, preliminary investigations suggested an AFM state for $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$, since a magnetic field $B \perp c$ of about 0.5 T depresses the transition temperature to zero. Application of pressure on Yb systems stabilizes its trivalent magnetic state and, therefore, reduces the Kondo energy scale. For $x = 0.27$ a Kondo temperature $T_K = 7.5$ K was found [24] which is about 1/3 of that of pure YbRh_2Si_2 .

In this letter, we present the low temperature properties of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ by means of ac-susceptibility, specific heat and magnetization in zero field and with external magnetic fields applied along the crystallographic c -direction. Surprisingly, we find the second order phase transition at $T_C = 1.30$ K to be ferromagnetic. This is shown by the high value of the susceptibility peak, the hysteresis in magnetization as well as by the behavior of the field-dependent susceptibility and specific heat. A small spontaneous magnetic moment $0.1 \mu_B/\text{Yb}$ is determined. A second, lower-lying phase transition T_L could not be detected down to the lowest achieved temperature of 0.02 K. Such a T_L transition exists at lower Co concentration and for pure YbRh_2Si_2 at pressures below 4 GPa (cf. Fig. 1a). Since preliminary results suggest that for $x < 0.27$ the phase below T_N is indeed of AFM type, one might find the phase below T_L to be FM.

For all measurements we used the same single crystal which was grown by indium flux technique [24]. The size of the sample is approximately 1.4 mm x 1.4 mm x 0.5 mm where the last value is the length along the c -direction. The cobalt content $x = 0.27(1)$ was measured by energy-dispersive x-ray spectroscopy. The low temperature measurements were performed in Oxford Instruments ^3He - ^4He -dilution refrigerators in a temperature range $0.02 \leq T \leq 5$ K and magnetic fields up to 1 T. The magnetization was measured with a Faraday magnetometer [33] and the ac-susceptibility with a standard susceptometer

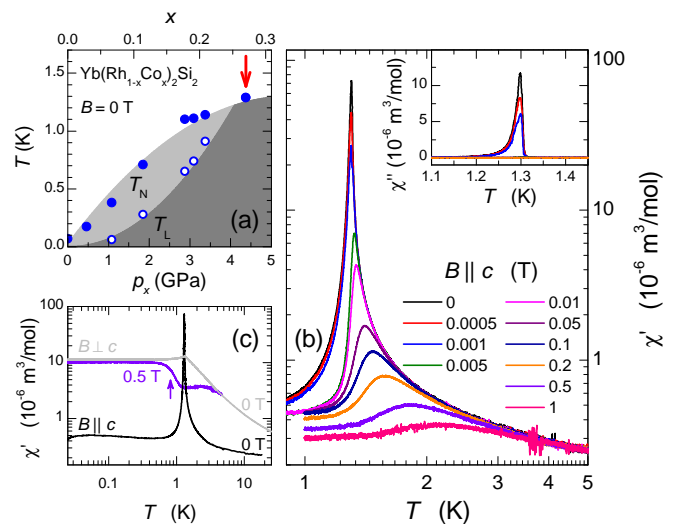


Figure 1: (Color online) (a) Excerpt of the magnetic phase diagram of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ from Ref. [24]. Open and closed circles correspond to the transition temperatures T_L and T_N , respectively. For $x \leq 0.12$, the data were taken from susceptibility measurements [25], while for larger Co content the data points were obtained by specific heat measurements [24]. The red arrow marks the position of the present system, where we observe FM order. The lower axis marks the corresponding hydrostatic pressure p_x on YbRh_2Si_2 (cf. Ref. [24]). (b) Ac-susceptibility $\chi'(T)$ of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ in different magnetic fields with $B \parallel c$. At zero field a sharp peak is observed at $T_C = 1.30$ K. With increasing magnetic field the peak decreases rapidly and its position shifts towards higher temperatures. Inset: Temperature dependence of the imaginary part of the susceptibility $\chi''(T)$. (c) Ac-susceptibility $\chi'(T)$ of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ parallel (black) and perpendicular (gray) to the c -direction in zero magnetic field. A magnetic field $B \perp c = 0.5$ T (violet) suppresses the phase transition towards lower temperatures (marked by arrow).

with a modulation field $B_{rms} = 11 \mu\text{T}$. The absolute values were obtained by matching the data to measurements done in a commercial Quantum Design SQUID Vibrating Sample Magnetometer. The specific heat was measured with a semi-adiabatic heat pulse technique [34] and a Quantum Design Physical Properties Measurement System.

Figure 1b shows the ac-susceptibility as a function of temperature in various magnetic fields. At zero field a sharp maximum is observed at $T_C = 1.30$ K with a huge absolute value of $73 \cdot 10^{-6} \text{ m}^3/\text{mol}$. With the molar volume $V_{mol} = 46.7 \cdot 10^{-6} \text{ m}^3/\text{mol}$, we obtain the dimensionless volume susceptibility $\chi_{vol} = \chi'/V_{mol} = 1.5$ which corresponds to a lower limit of the intrinsic susceptibility because of demagnetization effects due to the sample shape. However, the large value of χ_{vol} points to a FM phase transition, or at least a phase transition with a FM component along the c -axis. The application of an external static magnetic field strongly influ-

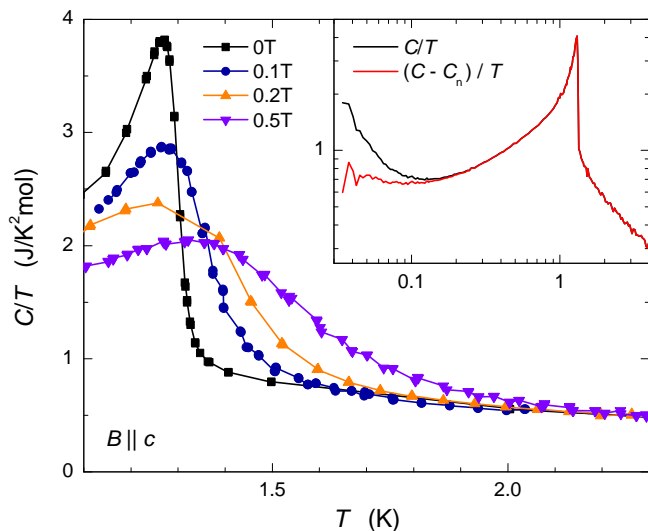


Figure 2: (Color online) Specific heat C/T vs T of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ in different magnetic fields. At $B = 0$, C/T features a mean-field-like anomaly at $T_C = 1.30$ K. Increasing B , the anomaly is shifted towards higher temperatures. Inset: The black curve shows the specific heat in zero field down to 0.03 K. The red curve shows the same data with the nuclear Schottky contribution subtracted.

ences the size of the peak. A tiny field $B = 0.001$ T reduces its size by more than a factor of 2. Increasing B , the maximum shifts towards higher temperatures as expected for the crossover temperature (which separates field-polarized and paramagnetic state) of a ferromagnet. The inset of figure 1b shows the imaginary part $\chi''(T)$ of the susceptibility. $\chi''(T)$ exhibits a clear feature within the critical region between 1.15 and 1.35 K at very low fields indicating the presence of dissipation (non-zero hysteresis), characteristic of ferromagnets. At fields above 0.01 T the imaginary part vanishes. The susceptibility of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ is reminiscent to the situation in YbNi_4P_2 which is located close to a FM-QCP [35]. Interestingly, in both systems the magnetic hard direction, which has a tiny susceptibility at enhanced temperatures, eventually becomes much larger slightly above T_C . This can be seen by comparing our data with the susceptibility for $B \perp c$, which is shown in figure 1c. Moreover, $\chi'(T)$ for $B \perp c = 0.5$ T demonstrates the suppression of the phase transition towards lower temperatures unlike the crossover for the $B \parallel c$ case. The phase diagram in terms of specific heat, magnetization, ac-susceptibility and electrical resistivity for $B \perp c$ will be published in a forthcoming paper [36].

The specific heat of $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ is depicted in Fig. 2. At zero field, $C(T)/T$ shows a mean-field-like transition at $T_C = 1.30$ K in agreement with the peak temperature found in $\chi'(T)$. The application of a magnetic field shifts T_C towards higher temperatures and the maximum broadens as expected for a FM phase transi-

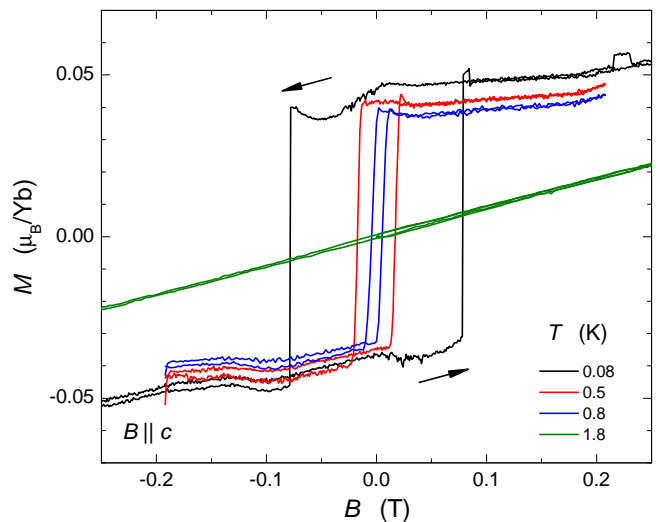


Figure 3: (Color online) Isothermal magnetization curves for different temperatures with $B \parallel c$. The field axis is shifted by 0.008 T in order to compensate for the remanent field due to the magnet gradient field. The arrows indicate the field sweep direction. The spontaneous magnetization is very small, and the obtained total signal is close the magnetometer's resolution limit.

tion. In the inset of Fig. 2 the low temperature specific heat (black curve) is shown at $B = 0$. The subtraction of the quadrupolar contribution of the Yb nucleus gives only a tiny correction to the data [29, 37]. A more important contribution can be ascribed to the magnetic moment of the $4f$ -electrons which creates a magnetic field at the Yb nucleus. This leads to an additional Schottky splitting of the nuclear dipole moment which gives rise to a T^{-2} contribution to $C(T)/T$ even at zero field. We suppose that the electronic contribution becomes constant at lowest temperatures and that the low temperature divergence is due to nuclear Schottky splitting induced by the spontaneous magnetization. In order to obtain a constant $C(T)/T$, we subtracted the nuclear Schottky contribution from our specific heat data by adjusting the spontaneous moment of the $4f$ -electrons. For this we used the hyperfine coupling constant determined from NMR measurements [38] and the size of the nuclear dipole moment [37]. The result is depicted by the red curve in the inset of Fig. 2 which exhibits a significant deviation from the raw data below 0.2 K. From this procedure we can extract a $4f$ magnetic moment of $0.20(5) \mu_B/\text{Yb}$.

Figure 3 illustrates the magnetization $M(B)$ as a function of magnetic field at various temperatures. At the lowest achieved temperature of 0.08 K a clear hysteresis is observed with a coercive field of about 0.08 T and a tiny spontaneous magnetic moment $0.05 \mu_B/\text{Yb}$. The hysteresis shrinks with increasing temperature and disappears for $T \geq T_C$. The size of the magnetic moment is smaller than the value from the specific heat measure-

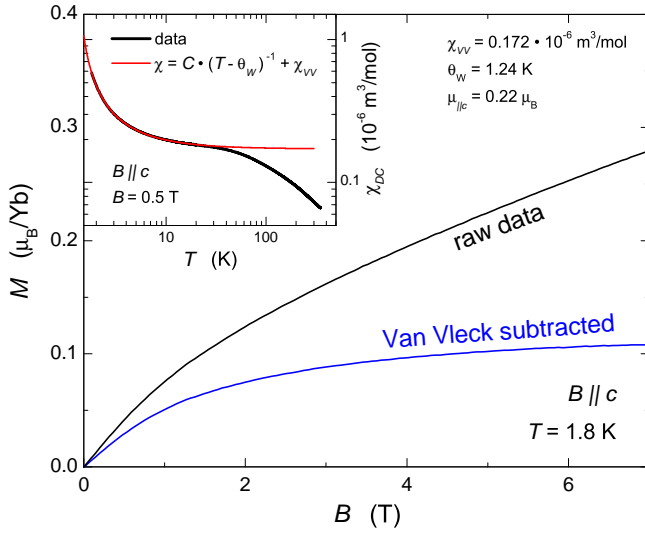


Figure 4: (Color online) Magnetization as a function of magnetic field $B \parallel c$ at $T = 1.8$ K. The Van Vleck contribution $\chi_{VV} = 0.172 \cdot 10^{-6} \text{ m}^3/\text{mol}$ was subtracted from the raw data, leaving a saturation magnetization at 7 T of about $0.11 \mu_B/\text{Yb}$. Inset: data of $\chi_{DC} = M/B$ between 2 and 300 K. Below 20 K a Curie-Weiss fit (red curve) was carried out as described in the main text.

ments. This might be due to a complex magnetic structure (e.g., spiral) with the total $4f$ magnetic moment being larger than its component in c -direction.

In Fig. 4 the magnetization M is plotted as a function of B above T_C at $T = 1.8$ K (black curve). Up to a field of 7 T the curve does not saturate which is due to a considerable Van Vleck contribution. To determine this contribution we performed a measurement of $\chi_{DC} = M/B$ as a function of T in an external magnetic field of 0.5 T. The data are shown in the inset of Fig. 4. The broad shoulder above 20 K corresponds to the first excited doublet of the $J = 7/2$ multiplet of the Yb^{3+} atom split by the crystalline electric field. We have fitted the region below 20 K with the function $\chi = C/(T - \Theta_W) + \chi_{VV}$ where C is Curie's constant, $\Theta_W = 1.24$ K is the Weiss temperature and $\chi_{VV} = 0.172 \cdot 10^{-6} \text{ m}^3/\text{mol}$ is the constant Van Vleck term. From the Curie's constant we can calculate the corresponding magnetic moment $0.22 \mu_B$ along the c -direction. This value is in nice agreement with the saturation magnetic moment of $0.25(5) \mu_B/\text{Yb}$ which is expected for the doublet ground state from electron-spin-resonance (ESR) results on $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$. There, a g -factor of $g = 0.5(1)$ was determined at $T = 5$ K along the c -direction [39]. In the main panel of Fig. 4 we have subtracted the Van Vleck contribution χ_{VV} from $M(B)$ and obtained the blue curve which seems to saturate above 7 T at about $0.11 \mu_B/\text{Yb}$. This value is substantially smaller than the values obtained from the Curie-Weiss fit and the ESR results, respectively. We ascribe this to the Kondo screening which still exists at $T = 1.8$ K

($< T_K = 7.5$ K) and 7 T ($< B_K = (k_B/g\mu_B)T_K \approx 22$ T).

To summarize, we have shown that $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ is a ferromagnet with a Curie temperature $T_C = 1.30$ K and a remanent magnetization along the crystallographic c -direction. This is evidenced by the huge absolute values of the ac-susceptibility, the hysteresis in the magnetization isotherms at $T < T_C$, and by the evolution of the transition in magnetic field. A spontaneous magnetic moment of $0.05 \mu_B/\text{Yb}$ has been measured along the c -direction in the FM phase, which is smaller than the value estimated from specific heat ($0.20(5) \mu_B/\text{Yb}$) which might be due to a complex magnetic structure. Neutron diffraction experiments were also performed on $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$, but, so far, no magnetic structure could be resolved below T_C . As a matter of fact, this is expected for a magnetic ordered moment of the order of $0.2 \mu_B$, which is below the resolution limit of common neutron diffractometers. The negative neutron diffraction results also suggest that, if an AFM component is present at all, it must be in the same order of magnitude. Above T_C , we recognized the c -direction to be the magnetic hard axis and determined a magnetic moment of $0.22 \mu_B$ from a Curie-Weiss fit below 20 K. After subtracting the Van Vleck contribution, the magnetization results taken at 1.8 K reveal $0.11 \mu_B/\text{Yb}$ at 7 T, which proves that the Kondo effect is operating. Our discovery opens the question whether the low-lying phase below T_L which is found for small Co concentrations in $\text{Yb}(\text{Rh}_{1-x}\text{Co}_x)_2\text{Si}_2$ or in the pure YbRh_2Si_2 (under pressure) is ferromagnetic, too. This might imply a field-induced FM QCP at $B_L < B_N$.

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